

# Imaging quasi-particle wavefunctions in quantum dots via tunneling spectroscopy

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We show that in quantum dots the physical quantities probed by local tunneling spectroscopies, namely the quasi-particle wavefunctions of interacting electrons, can considerably deviate from their single-particle counterparts as an effect of Coulomb correlation. From the exact solution of the few-particle Hamiltonian for prototype dots, we find that such deviations are crucial to predict wavefunction images at low electron densities or high magnetic fields.

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Current single-electron tunneling spectroscopies in semiconductor quantum dots (QDs) [1, 2, 3] may provide spectacular images of the QD wavefunctions, both in real [4, 5, 6] and reciprocal space [7, 8, 9]. The measured intensities have been generally attributed to the probability densities of ground or excited single electron states occupying the dot. As pointed out by Wibbelhoff *et al.* [9], however, the role of other electrons filling the dot may actually be relevant. Indeed, QDs can be strongly interacting objects with a completely discrete energy spectrum, which in turn depends on the number of electrons,  $N$  [1, 3]. Therefore, orbitals can be ill-defined, losing their meaning due to interaction. Also, it is unclear how many electrons one should take into account to calculate the total density of states, as a particle tunnels into a QD filled with  $N$  electrons. In this Letter we thus address the following basic questions: What are the physical quantities that are actually probed by scanning tunneling microscopies (STM) [4, 5, 6] or magneto-tunneling spectroscopies [7, 8, 9] of QDs? How do they depend on interactions? Can they deviate from the common single-particle picture in physically relevant regimes? If only one many-body state is probed at time, then the signal is proportional to the probability density of the *quasi-particle* (QP) being injected into the interacting QD. We demonstrate that the QP density dramatically depends on the strength of correlation inside the dot, and predict the wavefunction mapping to be a useful experimental tool to image QPs, both in direct and reciprocal space.

The imaging experiments, in their essence, measure quantities directly proportional to the probability for transfer of an electron through a barrier, from an emitter, where electrons fill in a Fermi sea, to a dot, with completely discrete energy spectrum. In multi-terminal setups one can neglect the role of electrodes other than the emitter, to a first approximation. The measured quantity can be the current [4, 7], the differential conductance [5, 6, 8, 10], or the QD capacitance [9, 11], while the emitter can be the STM tip [4, 5, 6], or a  $n$ -doped GaAs contact [7, 8, 9, 10, 11], and the barrier can be the vacuum [4, 5, 6] as well as a AlGaAs spacer [7, 8, 9, 10, 11].

According to the seminal paper by Bardeen [12], the

transition probability (at zero temperature) is given by the expression  $(2\pi/\hbar) |\mathcal{M}|^2 n(\epsilon_f)$ , where  $\mathcal{M}$  is the matrix element and  $n(\epsilon_f)$  is the energy density of the final QD states. The common wisdom would predict the probability to be proportional to the total density of QD states at the resonant tunneling energy,  $\epsilon_f$ , possibly space-resolved since  $\mathcal{M}$  would depend on the resonant QD orbital [13]. To proceed, let us assume that electrons from the emitter access through the barrier a single QD at a sharp resonant energy, corresponding to a unique, well defined many-body QD state, and reconsider the transition matrix element  $\mathcal{M}_{\mathbf{k},N}$  for transfer of an electron from emitter to QD.  $\mathcal{M}_{\mathbf{k},N}$  is given by [recasting Eqs. (6-7) of Ref. 12 in second quantized form]:

$$\mathcal{M}_{\mathbf{k},N} \propto \langle \{k\}, N-1 | \hat{\mathcal{M}} | \{k^*\}, N \rangle, \\ \hat{\mathcal{M}} = \frac{\hbar^2}{2m^*} \int \left[ \hat{\Psi}^\dagger \frac{\partial \hat{\Psi}}{\partial z} - \frac{\partial \hat{\Psi}^\dagger}{\partial z} \hat{\Psi} \right] \delta(z_{\text{bar}} - z) d\tau. \quad (1)$$

Here  $|\{k\}, N-1\rangle$  and  $|\{k^*\}, N\rangle$  are two many-particle states of the entire system of similar energies, with  $N-1$  and  $N$  interacting electrons in the QD, respectively, and the remaining  $N_{\text{tot}} - N + 1$  and  $N_{\text{tot}} - N$  electrons, respectively, in the emitter. The fixed coordinate along the tunneling direction  $z$  appearing in (1),  $z_{\text{bar}}$ , can be everywhere in the barrier, and  $d\tau$  is the infinitesimal volume element. The fermionic field operator  $\hat{\Psi}(\mathbf{r})$ , destroying an electron at position  $\mathbf{r} \equiv (\boldsymbol{\rho}, z)$ , can be expanded over the basis of emitter and QD single-particle states,  $\phi_{\mathbf{k}}$  and  $\phi_{\alpha}$ , respectively [14]:  $\hat{\Psi}(\mathbf{r}) = \sum_i \phi_i(\mathbf{r}) \hat{c}_i$ , where  $i = \mathbf{k}, \alpha$  and we take unitary volume normalization. We omit spin indexes and summations for the sake of simplicity. We assume that electrons in the emitter do not interact and are associated to the two sets of quantum numbers  $\{k\}$  and  $\{k^*\}$ , respectively, which differ in the occurrence of the index  $\mathbf{k}$  labeling the electron which leaves the emitter and tunnels to the QD as  $|\{k\}, N-1\rangle$  evolves to  $|\{k^*\}, N\rangle$ . Moreover, we assume for convenience that the  $xy$  and  $z$  motions of electrons are separable, and that electrons in the QD all occupy the same confined single-particle state along  $z$ ,  $\chi_{\text{QD}}(z)$ , namely  $\phi_{\alpha}(\mathbf{r}) = \phi_{\alpha}(\boldsymbol{\rho}) \chi_{\text{QD}}(z)$ . Under these conditions we may factorize the matrix element as

$\mathcal{M}_{\mathbf{k},N} \propto T_{\mathbf{k}} M_{\mathbf{k},N}$ , with

$$T_{\mathbf{k}} = \frac{\hbar^2}{2m^*} \left[ \chi_{\mathbf{k}}^*(z) \frac{\partial \chi_{\text{QD}}(z)}{\partial z} - \chi_{\text{QD}}(z) \frac{\partial \chi_{\mathbf{k}}^*(z)}{\partial z} \right]_{z=z_{\text{bar}}}, \quad (2)$$

where  $\chi_{\mathbf{k}}(z)$  is the emitter state along  $z$  evanescent in the barrier,  $\phi_{\mathbf{k}}(\mathbf{r}) = \phi_{\mathbf{k}}(\boldsymbol{\varrho}) \chi_{\mathbf{k}}(z)$ , and

$$M_{\mathbf{k},N} = \sum_{\alpha} \int \phi_{\mathbf{k}}^*(\boldsymbol{\varrho}) \phi_{\alpha}(\boldsymbol{\varrho}) d\boldsymbol{\varrho} \langle \{k\}, N-1 | \hat{c}_{\mathbf{k}}^{\dagger} \hat{c}_{\alpha} | \{k^*\}, N \rangle.$$

Eventually assuming that the many-body states can be factorized into an emitter and a QD part, we obtain

$$M_{\mathbf{k},N} = \int \phi_{\mathbf{k}}^*(\boldsymbol{\varrho}) \varphi_{\text{QD}}(\boldsymbol{\varrho}) d\boldsymbol{\varrho}, \quad (3)$$

where  $\varphi_{\text{QD}}(\boldsymbol{\varrho})$  is the quasi-particle (QP) wavefunction of the interacting QD system [15]:

$$\varphi_{\text{QD}}(\boldsymbol{\varrho}) = \langle N-1 | \hat{\Psi}(\boldsymbol{\varrho}) | N \rangle. \quad (4)$$

Results (3-4) are the key for predicting wavefunction images both in real and reciprocal space. In STM,  $\phi_{\mathbf{k}}(\boldsymbol{\varrho})$  is the localized tip wavefunction; if we ideally assume it point-like and located at  $\boldsymbol{\varrho}_0$  [13], i.e.  $\phi_{\mathbf{k}}(\boldsymbol{\varrho}) \approx \delta(\boldsymbol{\varrho} - \boldsymbol{\varrho}_0)$ , then the signal intensity is proportional to  $|\varphi_{\text{QD}}(\boldsymbol{\varrho}_0)|^2$ , which is the usual result of the one-electron theory [6, 13], provided the ill-defined QD orbital is replaced by the QP wavefunction unambiguously defined by Eq. (4). In magneto-tunneling spectroscopy, the emitter in-plane wavefunction is a plane wave,  $\phi_{\mathbf{k}}(\boldsymbol{\varrho}) = e^{i\mathbf{k} \cdot \boldsymbol{\varrho}}$ , and the matrix element (3) is the Fourier transform of  $\varphi_{\text{QD}}$ ,  $M_{\mathbf{k},N} = \varphi_{\text{QD}}(\mathbf{k})$ . Again, we generalize the standard one-electron result by substituting  $\varphi_{\text{QD}}(\mathbf{k})$  for the QD orbital [then Eqs. (3-2) coincide with (A1-A2) of Ref. 8]. Note that  $M_{\mathbf{k},N}$  is the relevant quantity also for intensities in space-integrated spectroscopies probing the QD addition energy spectrum [10, 11]. Consistently, in the non-interacting case,  $\varphi_{\text{QD}}(\boldsymbol{\varrho})$  reduces to the highest occupied one-electron orbital  $\phi_{\alpha}(\boldsymbol{\varrho})$  [13]: in this limit an electron tunnels from the emitter to the orbital  $\phi_{\alpha}(\boldsymbol{\varrho})$  which resonates at the Fermi energy, with  $|N\rangle = \hat{c}_{\alpha}^{\dagger} |N-1\rangle$ . The latter regime probably corresponds to most of the existing experimental evidence [4, 5, 6, 7, 8, 9]. However, it is interesting to analyze realistic scenarios that deviate from the one-electron picture.

Therefore, we study  $\varphi_{\text{QD}}(\boldsymbol{\varrho})$  in a paradigmatic interacting case, and consider a few electrons in a two-dimensional harmonic trap, which was proven to be an excellent model for different experimental setups [3]. The QD effective-mass Hamiltonian is

$$H = \sum_i H_0(i) + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{\kappa |\boldsymbol{\varrho}_i - \boldsymbol{\varrho}_j|}, \quad (5)$$

with

$$H_0(i) = \frac{1}{2m^*} \left[ \mathbf{p} - \frac{e}{c} \mathbf{A}(\boldsymbol{\varrho}) \right]^2 + m^* \omega_0^2 \varrho^2 / 2. \quad (6)$$

Here  $\kappa$  is the static relative dielectric constant of the host semiconductor, and  $\mathbf{A}(\boldsymbol{\varrho})$  is the vector potential ( $\mathbf{A} = \mathbf{B} \times \boldsymbol{\varrho} / 2$ ) associated with a static and uniform magnetic field  $B$  along  $z$ , which reduces the cylindrical spatial symmetry group of the system from  $D_{\infty h}$ , at  $B = 0$ , to  $C_{\infty h}$ , when  $B \neq 0$ , making it chiral. The QD wavefunction has an azimuthal quantum number  $m$ ,  $\varphi_{\text{QD}}(\boldsymbol{\varrho}) = \varphi_{\text{QD}}(\varrho) e^{im\varphi}$ , which is fixed by the total angular momenta  $M$  of  $|N\rangle$  and  $|N-1\rangle$ ,  $m = M_N - M_{N-1}$ , and can be expanded over the basis of Fock-Darwin (FD) orbitals  $\varphi_{nm}(\boldsymbol{\varrho})$  [1], eigenstates of the single-particle Hamiltonian (6):  $\varphi_{\text{QD}}(\boldsymbol{\varrho}) = \sum_{n=0}^{\infty} a_n \varphi_{nm}(\boldsymbol{\varrho})$ , where  $n$ 's are radial quantum numbers and  $a_n$  coefficients to be determined. We solve numerically the few-body problem of Eq. (5), for the ground state at different  $N$ 's, by means of the configuration interaction (CI) method [16], where  $|N\rangle$  is expanded in a series of Slater determinants built by filling in the FD orbitals with  $N$  electrons, and consistently with symmetry constraints [16]. Then, we evaluate the matrix element (4), and find the values of  $a_n$  for a truncated FD basis set.

There are two ways of artificially tuning the strength of Coulomb correlation in QDs: one is to dilute electron density, and the other is to turn on  $B$ . In both cases, at low enough densities or strong enough fields, electrons pass from a “liquid” phase, where low-energy motion is equally controlled by kinetic and Coulomb energy, to a “crystallized” phase, reminiscent of the Wigner crystal in the bulk, where electrons are localized in space and arrange themselves in a geometrically ordered configuration such that electrostatic repulsion is minimized [3, 20].

We first consider reducing the density at  $B = 0$ . The typical QD lateral extension is given by the characteristic dot radius  $\ell_{\text{QD}} = (\hbar / m^* \omega_0)^{1/2}$ ,  $\ell_{\text{QD}}$  being the mean square root of  $\varrho$  on the FD lowest-energy level  $\varphi_{00}$ . As we keep  $N$  fixed and increase  $\ell_{\text{QD}}$ , the Coulomb-to-kinetic energy ratio  $\lambda = \ell_{\text{QD}} / a_{\text{B}}^*$  [ $a_{\text{B}}^* = \hbar^2 \kappa / (m^* e^2)$  is the effective Bohr radius of the dot] [18] increases as well, driving the system into the “Wigner” regime [21]. As a rough indication, consider that for  $\lambda \approx 2$  or lower the electronic ground state is liquid, while above  $\lambda \approx 4$  electrons form a “crystallized” phase [18].

Figure 1 shows  $\varphi_{\text{QD}}$  vs.  $\varrho$ , as up to six electrons are successively injected into a “liquid” QD with a realistic density of  $\lambda = 2$  [10]. The QD filling sequence is well known [10, 11], in analogy with the Aufbau principle of atomic physics: in the independent-electron picture ( $\lambda = 0$ , dashed lines),  $\varphi_{\text{QD}}$  is the highest-energy occupied orbital which is filled by the electron added to the dot. However, Coulomb correlation significantly spreads the wavefunction (solid lines) and moves the QP peak towards the QD edge. The spreading is caused by the increase of weights  $a_n$  of high-energy FD orbitals, as interaction is turned on; nevertheless, the behavior of  $\varphi_{\text{QD}}$  around  $\varrho \approx 0$  is dictated by its angular dependence,  $\varphi_{\text{QD}}(\varrho) \propto \varrho^{|m|}$ , while it decays like  $\exp(-\varrho^2 / 2\ell_{\text{QD}}^2)$  as

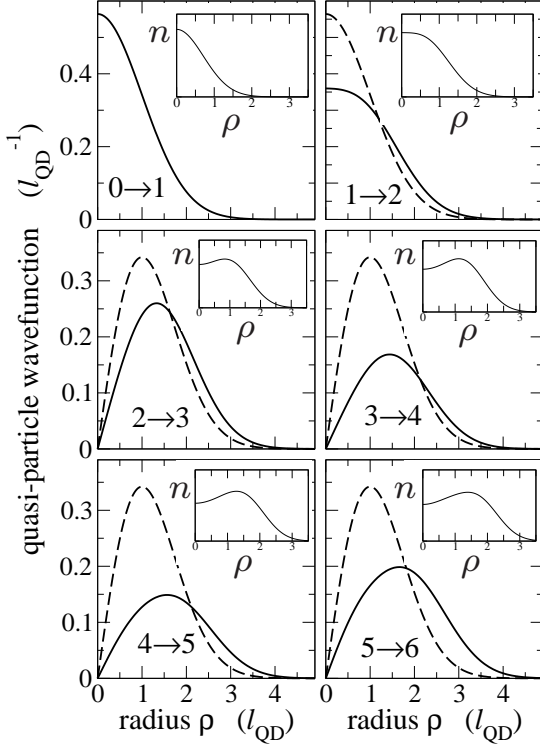


FIG. 1: Quasi-particle wavefunction (solid line) vs.  $\varrho$  for different  $(N-1) \rightarrow N$  transitions, with  $\lambda = 2$ . The dashed line represents the non-interacting orbital ( $\lambda = 0$ ). The ground states are  $(M, S) = (0, 1/2), (0, 0), (1, 1/2), (0, 1), (1, 1/2), (0, 0)$ , for  $N$  going from 1 to 6, respectively.  $S_z = 0$  ( $S_z = 1/2$ ) if  $N$  is even (odd). The norm of the  $\lambda = 2$  wavefunction is 1, 0.84, 0.84, 0.40, 0.37, 0.73, respectively. Insets: Total ground-state charge densities  $n(\varrho)$  (arb. units) for  $N$  going from 1 (top left) to 6 (right bottom). The length unit is  $\ell_{\text{QD}}$ .

$\varrho \rightarrow \infty$ . The QP amplitude is strongly suppressed in the  $(N-1) \rightarrow N$  tunneling processes involving the  $N = 4$  open-shell ground state, with respects to other additions (Fig. 1). This is a spin-blockade effect, since the total spin,  $S$ , is maximum at  $N = 4$  ( $S = 1$  according to Hund's rule [10]), and we assume that its  $z$ -component is zero,  $S_z = 0$ . Besides, the general trend is that the QP wavefunction norm, hence the integrated experimental signal, diminish as  $N$  and  $\lambda$  increase (see also Fig. 2).

Note that the interpretation of tunneling spectroscopy in terms of the total density,  $n(\varrho) = \langle N | \hat{\Psi}^\dagger(\varrho) \hat{\Psi}(\varrho) | N \rangle / N$ , is inconsistent with our point of view, as it is seen by comparing QP wavefunctions of Fig. 1 with total densities for the corresponding  $N$ -electron states (insets). While total densities and QP probabilities resemble each other up to the addition of the second electron, after the third electron tunnels into the dot they can be clearly discriminated in the laboratory: QP probabilities have a strong angular dependence (hybridizing degenerate states with  $\pm m$ ) and a node at the QD center, while total densities are approximately

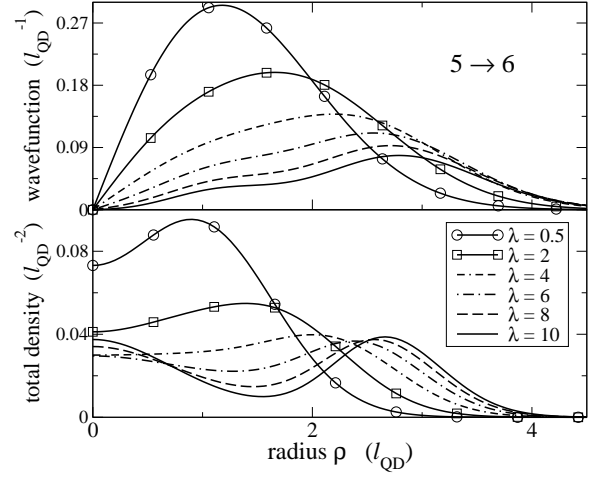


FIG. 2: Top: Quasi-particle wavefunction vs.  $\varrho$  for different values of  $\lambda$ , as the 6-th electron tunnels into the QD. The wavefunction norm, for  $\lambda$  going from 0.5 to 10, is 0.97, 0.73, 0.48, 0.32, 0.22, 0.15, respectively. Bottom: Six-electron total density  $n(\varrho)$  vs.  $\varrho$ . The ground states for  $N = 5, 6$  are  $(M, S) = (1, 1/2), (0, 0)$ , respectively, for all  $\lambda$ .

circular (exactly, for  $N = 4, 6$ ) and filled.

As one reduces the density, the appearance of QP wavefunctions dramatically changes. In Fig. 2 we study the injection of the 6th electron, as  $\lambda$  goes from 0.5 up to 10. Note that  $\lambda = 0.5, 2, 4$  (equivalent to GaAs lateral confinement energies  $\hbar\omega_0 = 47, 3.0, 0.74$  meV, respectively), typically correspond to different experimental QD devices, such as self-assembled [8, 22], vertical mesa etched [10], and 2DEG-depletion QDs [11]. A six-electron Wigner molecule forms for  $\lambda > 4$ , with one electron localized at the QD center, and the remaining five arranged on an outer ring, at the vertices of a regular pentagon [18, 23, 24]. The crystallization is clearly seen in the bottom panel of Fig. 2, where, as  $\lambda$  increases, the total density develops one peak at  $\varrho = 0$ , for the central electron, and another one, close to  $\varrho = 3\ell_{\text{QD}}$ , for the outer ring. Similarly, the five-electron molecule is a hollow pentagon [18, 23]. In the top panel of Fig. 2 we see that the QP wavefunction is strongly affected by electron localization: while for  $\lambda < 4$  it somehow resembles the non-interacting FD orbital  $(n, m) = (0, -1)$ , being spread uniformly across the dot, for  $\lambda > 4$  it develops a well formed peak close to the outer-ring position. The QP weight in the region inside the ring is strongly depleted, eventually appearing as a shoulder of the main peak. We conclude that, in the crystallized phase, the 6th electron can only enter the external ring, with negligible probability of being located into the center. For smaller  $N$ , we find that electrons just enter the outer ring, since the pertinent geometrical phases are hollow regular polygons [25].

We now come to the effect of a strong magnetic field parallel to the tunneling direction  $z$ . As  $B$  increases, the

$(N-1) \rightarrow N$	$\nu = 1$	$\nu = 1/2$	$\nu = 1/3$	$\nu = 1/4$	$\nu = 1/5$
$1 \rightarrow 2$	1.00	1.00	0.500	0.707	0.250
$2 \rightarrow 3$	1.00	0.430	0.336	0.190	0.106
$3 \rightarrow 4$	1.00	0.520	0.270	0.201	0.0649
$4 \rightarrow 5$	1.00	0.158	0.239	0.0650	0.0507
$5 \rightarrow 6$	1.00	0.294	0.210	0.0541	0.0274

TABLE I: Absolute value of the modulation coefficient  $|a_0|$  of the quasi-particle wavefunction  $\varphi_{\text{QP}}(\mathbf{q}) = a_0 \varphi_{0m}(\mathbf{q})$ , where  $m = (N-1)/\nu$ , for different  $(N-1) \rightarrow N$  tunneling processes and filling factors  $\nu$ .

kinetic energy is quenched, Landau bands of almost degenerate FD levels being formed.  $M$  increases due to Coulomb repulsion, since the higher  $m$ , the outer the FD orbital [26]. In correspondence of “magic” values of  $M$ , the ground state turns out to be particularly stable [26]: this family of incompressible [27] states has been variously regarded as reminiscent of fractional quantum Hall effect (FQHE) states in two-dimensional electron layers [3], or as a collection of Wigner molecules [28].

In analogy with FQHE, it is convenient to introduce the filling factor  $\nu$ , defined as  $\nu = N(N-1)/2M$ , and to consider only FD levels in the lowest Landau band and full spin-polarization, which turns out to be a reasonable approximation at high  $B$  [3]. In realistic situations, there are significant  $B$ -ranges where  $\nu$  is constant as  $N$  is changed [3, 29]. At  $\nu = 1$ , the interacting states are maximum density droplets [3, 30], namely incompressible disks of almost uniform density,  $|N\rangle = \prod_{m=0}^{N-1} \hat{c}_{0m}^\dagger |0\rangle$ , and  $\varphi_{\text{QP}}$  is simply the highest occupied FD state,  $\varphi_{0N-1}$ , located at the edge of the dot, which is being filled by the tunneling electron, with  $a_n = \delta_{n0}$ :

$$\varphi_{\text{QP}}(\mathbf{q}) = \varphi_{n=0, m=N-1}(\mathbf{q}). \quad (7)$$

Equation (7) is a remarkable result: while the total electron density is a uniform disk, the measured squared modulus of QP wavefunction will be an annulus of the same radius as the charge distribution. If  $\nu < 1$ , the wavefunction will be still proportional to the FD orbital  $\varphi_{n=0, m}$ , with  $m = (N-1)/\nu$  and  $a_0 \neq 1$ , namely  $\varphi_{\text{QP}}(\mathbf{q}) = a_0 \varphi_{n=0, m}(\mathbf{q})$ . The only effect of strong correlation in these regimes is to modulate the amplitude of the non-interacting wavefunction via the coefficient  $a_0$ . Table I shows the calculated values of  $a_0$  for various tunneling processes at particularly stable filling factors. Except for some cases,  $|a_0|$  monotonously decreases as  $\nu$  diminishes or as  $N$  increases. E.g., at  $\nu = 1/5$   $|a_0|$  is reduced by two order of magnitudes with respect to  $\nu = 1$ , when the 6th electron enters the dot. Table I shows that interaction enforces very effectively the orthogonality of incompressible states [31], and therefore we expect that, as a high field component is applied parallel to  $z$ , tunneling is strongly suppressed by the reduction of the matrix

element  $M_{\mathbf{k}, N}$  [Eq. (3)]: a purely many-body mechanism, the single-particle matrix element  $T_{\mathbf{k}}$  [Eq. (2)] being left unchanged by the field.

In conclusion, we have shown that quasi-particle wavefunctions of QDs are extremely sensitive to electron-electron correlation, and may differ from single-particle states in physically relevant cases. This result is of interest to predict the real- and reciprocal-space wavefunction images obtained by tunneling spectroscopies, as well as the intensities of addition spectra of QDs. Close comparison with experiment is not yet possible in the case of Ref. [9], where many dots are probed at once and the confinement is too strong. Promising samples are also those of Refs. [10, 11], allowing for access to a single dot and full control of  $N$ . We hope that our results will stimulate further experiments. We believe that our findings will be important also for other strongly confined systems, like e.g. nanostructures at surfaces [32].

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  - [16] For details on our CI approach see Ref. 17. Here we implemented a parallel version of our CI code, allowing for using a FD basis set at  $B = 0$  as large as 36 orbitals, and for diagonalizing matrices of linear dimensions up to  $\approx 10^6$ . As a convergence test, we could accurately reproduce Quantum Monte Carlo ground state energies up to  $\lambda = 8$  and  $N = 6$  [18]. Details will be given in Ref. 19.

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